IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Patent Application of:

          Koji KOYANAGI et al.

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For: SURFACTANT COMPOSITION		Examiner: A. LI

DECLARATION UNDER 37 C.F.R. § 1.132

Commissioner for Patents
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Sir:

     I, Hotaka Yamamuro, hereby declare as follows:

     I am one of the co-inventors of the invention as described and claimed in the above-identified patent application.

     I am a graduate of Tsukuba University and received my Master's degree in 1992, majoring in applied biochemistry.

     I have been employed by Kao Corporation since 1992. In 1992, I was a researcher involved in Performance Chemicals Research Laboratories with a responsibility of Chemical Admixtures for Concrete.

     The cationic polymer (C) of the present invention produces unexpectedly superior results. When the surfactant composition of the present invention is used for a slurry rheology modifier, a combination of the compounds (A) and (B) and the cationic polymer (C) produces such an excellent rheology modifying effect on even a slurry containing clay. Although not wanting to be limited by theory, the reason is as follows: Specifically, in the slurry in which clay exists, compound (A) adsorbs to the clay and there is therefore the case where the formation of string-like micelles which serve to modify rheology is inhibited. However, if the cationic polymer (C) exists like the case of the present invention, the adsorption of the cationic polymer (C) to clay can prevent compound (A) from adsorbing to clay because the cationic polymer (C) adsorbs to clay more easily than compound (A). Also,

the adsorption of the cationic polymer (C) to clay causes the coagulation of clay particles and also, the surface area of clay is reduced, ensuring that the amount of compound (A) to be adsorbed to clay can be reduced. As a result, string-like micelles (huge micelle associated body) produced by the compounds (A) and (B) are formed sufficiently and therefore, the original effects of the compounds (A) and (B) are maintained.

It is considered that the present invention can prevent compound (A) from adsorbing to clay by the aid of the cationic polymer (C) in the case where sufficient rheology modifying effect is not obtained because of the adsorption of compound (A) to clay, and therefore, rheology modification can be accomplished even in the case where substances having the ability to adsorb compound (A) are present besides the clay.

The claimed density of the cationic polymer (C) also produces unexpectedly superior results. As to the index of the ability to adsorb compound (A), it is preferable to apply the present invention to materials having a chemical equivalent of 0.1 meq or more (0.1 meq/100 g or more) to compound (A) per 100 g. Materials having a chemical equivalent of 1 to 10 meq/100 g in particular have a difficulty in obtaining an intended rheology modifying effect even if the amount of compound (A) to be added is significantly increased. Therefore, the present invention is preferable in such a case.

As further evidence, the following examples were carried out by myself or under my direct supervision. Test procedures and results are shown below.

As the slurry raw materials, the following materials were used:

(i) Hydraulic powder: Ordinary portland cement, density: 3.16 g/cm^3, Taiheiyo Cement (K.K.)

(ii) Compound (A): Hexadecyltrimethylammonium chloride/octadecyltrimethylammonium chloride = 50/50 (weight ratio) (used as an aqueous 29 wt.%) solution (viscosity at 20°C: 18 mPa·s))

(iii) Compound (B): Sodium p-toluenesulfonate (used as an aqueous 20 wt.% solution (viscosity at 20°C: 2.5 mPa·s))

(iv) Clay: Kasaoka Clay (Kanesan Kogyo (K.K.))

(v) Additive: as described in Table 1

The viscosity of an aqueous solution obtained by mixing an aqueous 29 wt.% solution of compound (A) with an aqueous 20 wt.% solution of compound (B) in a ratio of 50/50 was 200,000 mPa·s at 20°C.

For the preparation of a slurry, 400 g of cement, 20 g of clay, and 400 g of water containing compound (B) and the additives shown in Table 1 (containing 8 g (2 wt.% based on the total weight of water) of an aqueous solution of compound (B) were mixed by a hand mixer for 30 seconds. Then, 8 g (2 wt.% based on the total weight of water) of an aqueous solution of compound (A) was mixed in this mixture, followed by mixing by a hand mixer for 60 seconds.

With regard to the obtained slurry, its viscosity was measured just, 60 minutes, and 120 minutes after the slurry was prepared. The viscosity was measured at 20°C by using Viscotester (using No. 1 rotor) manufactured by Ryon. The results are shown in Table 1. As shown in Table 1, Comparative Product 1 is reduced largely in the viscosity of the slurry with time and Comparative Products 2 to 6 results in segregation of the slurry. In the case of the products of the present invention, on the other hand, the viscosities of all the slurries are stable over time and no segregation occurs. Here, the term "the segregation of the slurry" means that a part of water in the slurry is separated from powder and aggregate and floated on the upper layer of the slurry. Like EP 1266875 to Yamamuro, the Comparative Products do not contain cationic polymer (C) of the present invention.

Table 1

		Additive		Viscosity (mPa · s／20℃)		
		kind	weight %	After 0 minute	After 60 minutes	After 120 minutes
Product of the invention	1	Cationic polymer (1)	34.5	6100	7000	7500
	2	Cationic polymer (2)	34.5	7000	7800	6900
	3	Cationic polymer (3)	34.5	7400	7400	7400
	4	Cationic polymer (4)	34.5	7400	6800	6100
	5	Cationic polymer (5)	34.5	6500	6800	5500
	6	Cationic polymer (6)	34.5	6000	6500	6000
	7	Cationic polymer (7)	34.5	6000	6000	6000
	8	Cationic polymer (8)	34.5	6000	6000	6000
	9	Cationic polymer (1)	13.8	6000	6500	7100
Comparative product	1	Cationic compound	34.5	6000	4500	3000
	2	$CaCl_2 \cdot 2H_2O$	172.5	6000	1500	segregation
	3	$CaCl_2 \cdot 2H_2O$	69	4900	1500	segregation
	4	Polymer(1)	34.5	2000	segregation	segregation
	5	Polymer (2)	34.5	4000	9000	segregation
	6	None	—	2400	700	segregation

"Wt.%" of the additives in the table is the wt.% of the effective content of the additive based on the effective content of compound (A). Also, each polymer is selected from the following.

· Cationic polymer (1): Poly(diallyldimethylammonium chloride), manufactured by Aldrich, low-molecular weight product (weight average molecular weight 5000 to 20000 (indicated on a ravel), cation density: 6.13 meq/g (used in the state of an aqueous 40 wt.% solution)

· Cationic polymer (2): Poly(diallyldimethylammonium chloride), manufactured by Aldrich, weight average molecular weight 100,000 to 200,000 (indicated on a ravel), cation density: 6.19 meq/g (used in the state of an aqueous 20 wt.% solution)

· Cationic polymer (3): Poly(diallyldimethylammonium chloride), manufactured by Aldrich, weight average molecular weight 400,000 to 500,000 (indicated on a ravel), cation density: 6.15 meq/g (used in the state of an aqueous 20 wt.% solution)

· Cationic polymer (4): Polymethacryloyloxyethyldimethylethylammoniumethyl sulfate, weight average molecular weight 120,000, cation density: 3.63 meq/g (used in the state of an aqueous 36.5 wt.% solution)

· Cationic polymer (5): Diallyldimethylammonium chloride-SO$_2$ copolymer, weight average molecular weight 4,000, trade name: PAS-A-5 (Nittobo (K.K.)), cation density: 4.33 meq/g

· Cationic polymer (6): Trade name: Accurac 41 (Mitsui Saitech (K.K.), weight average molecular weight 40,000, cation density: 7.10 meq/g (used in the state of an aqueous 50 wt.% solution)

· Cationic polymer (7): Trade name: Accurac 35 (Mitsui Saitech (K.K.), weight average molecular weight 70,000, cation density: 7.11 meq/g (used in the state of an aqueous 50 wt.% solution)

· Cationic polymer (8): Trade name: Accurac 57 (Mitsui Saitech (K.K.), weight average molecular weight 250,000, cation density: 7.27 meq/g (used in the state of an aqueous 50 wt.% solution)

· Cationic compound: Tetramethylammonium chloride (reagent), molecular weight 109.6, cation density: 9.12 meq/g (effective content: 100%)

· Polymer (1): Carboxymethyl cellulose, trade name: CMC1190 (Daicel Chemical Industries, Ltd.)

· Polymer (2): Polyvinylpyrrolidone, trade name: K-60 (ISP TECHNOLOGIES INC.)

The cation density of the cationic polymer was measured (colloid titration) in the following manner.

First, the cationic polymer (may be a pure cationic polymer or may be put in a solution state) was dissolved in water adjusted to pH 3.0 by phosphoric acid. A toluidine blue indicator was added to the solution, which was then subjected to titration using a 1/400 N potassium polyvinylsulfate solution, provided that the point at which the color of the solution was changed was regarded as the end point. The cation density was found according to the following equation.

Cation density (meq/g) = $(1/400) \times f \times [(mL)/1000] \times 1000 \times 1/[(g) \times (\%)/100]$

where:

f: Factor of 1/400 N potassium polyvinylsulfate solution;

(mL): Amount of the potassium polyvinylsulfate solution to be added;

(g): Amount of the sample; and

(%): Concentration of the sample.

The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S. Code 1001 and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

By: _Hotaka Yamamuro_   Date: _6 / 18 / 2010_
      Hotaka Yamamuro